Table A1. Radiocarbon Samples and Locations

State	Collection Date	Latitude/Longitude Coordinates	Elevation (m)	Nearest City	Distance to City (km)	Height of Corn (m)	Species	Number of Samples Run	$\Delta^{14}C$ Value (‰)	STD of Multiple Obs.[1]
Alabama										
	7/18/2004	32°37.000′ N 86°35.905′ W	155	Claton	19	2.1	Zea mays	1	60.6	-
	7/18/2004	34°06.561′ N 86°52.850′ W	215	Cullman	2	2.1	Zea mays	2	61.1	0.4
	7/17/2004	32°20.265′ N 86°24.545′ W	55	Montgomery	0	1.8	Zea mays	1	56.9	-
Alaska										
	7/29/2004	63°9′ N 149°0′ W	410	Healy	16	-	Annual Plant	2	65.2	0.2
California										
	8/11/2004	37°55′54″ N 121°41′44″ W	25	Brentwood	5	1.2	Zea mays	1	37.2	-
	8/14/2004	33°12′38″ N 117°28′59″ W	15	Carlsbad	-	-	Zea mays	1	54	-
	8/14/2004	36°49′30″ N 119°42′10″ W	110	Clovis	-	-	Zea mays	2	55.3	2.5
	8/14/2004	33°57′12″ N 117°23′46″ W	250	Riverside	-	-	Zea mays	2	42.8	0.4
	8/26/2004	37°20′ N 122°23′ W	20	San Gregorio	3	1.2	Zea mays	2	67.1	-
Colorado										
	8/12/2004	38°28′41″ N 107°52′34″ W	1770	Montrose	-	-	Zea mays	4	64.2	3.2
	7/20/2004	40°37′31″ N 103°12′27″ W	1200	Sterling	6	1.8	Zea mays	1	67.5	-
Florida										
	~7/1/2004	26°55'46" N 82°02'44" W	-	Gainesville	-	-	Zea mays	2	55.8	4.1
Georgia										
	7/8/2004	34°56′06″ N 83°29′38.4″ W	585	Clayton	19	2.1	Zea mays	1	63.9	-
	7/17/2004	31°31′50″ N 83°50′07″ W	130	Sylvester	16	1.8	Zea mays	2	63.3	0.1
Hawaii										
	7/24/2004	20°01′23″ N 155°40′18″ W	815	Waimea	0	-	Zea mays	2	65.8	0.7
Idaho										

	7/31/2004	42°11′10″ N 111°24′04″ W	1835	Bloomington	0	1.5	Zea mays	1	67.3	-
	8/20/2004	42°48′43″ N 114°53′55″ W	900	Hagerman	-	2	Zea mays	1	65.7	-
	8/15/2004	43°48′ N 116°44′ W	735	Sand Hollow	-	1.8	Zea mays	2	67.4	2.2
Indiana										
	7/17/2004	38°13′26″ N 86°51′43″ W	165	Ferdinand	8	2.4	Zea mays	1	62.4	-
Kansas										
	7/17/2004	39°19′01″ N 96°58′99″ W	310	Manhattan	13	2.1	Zea mays	3	65.3	0.7
Kentucky										
	7/19/2004	37°43.370′ N 85°31.487’ W	170	Bardstown	5	2.1	Zea mays	1	60.8	-
	7/20/2004	38°11.421′ N 83°24.812′ W	230	Morehead	1	2.1	Zea mays	1	64.9	-
	7/19/2004	37°01.510′ N 86°09.143′ W	210	Smith's Grove	19	2.1	Zea mays	2	60.2	0.8
Maryland										
	7/21/2004	39°35.844′ N 77°37.974′ W	160	Beaver Creek	2	2.1	Zea mays	1	56.5	-
	7/22/2004	39°00′16″ N 76°52′31″ W	55	Greenbelt	4	0.8	Zea mays	1	52	-
	7/21/2004	39°42.892′ N 78°21.408′ W	285	Pinley Grove	3	2.1	Zea mays	2	58.9	2.4
Massachusetts										
	8/5/2004	41°69.850′ N 70°28.486′ W	10	Barnstable	3	1.8	Zea mays	1	58.4	-
	7/24/2004	41°33′05″ N 70°36′53″ W	5	Falmouth	3	0.5	Zea mays	1	54.9	-
	8/25/2004	42°29′15″ N 72°11′15″ W	330	Petersham	5	-	Zea mays	1	58.3	-
	7/26/2004	42°26′96″ N 71°80′89″ W	145	Worcester	16	1.8	Zea mays	2	62.1	2.9
Michigan										
	8/15/2004	45°35' N 84°34' W	1615	Cheboygan	11	1.3	Zea mays	1	64.8	-
	7/19/2004	44°18′44″ N 83°47′38″ W	270	Hale	7	0.9	Zea mays	1	68.9	-
Minnesota										
	8/19/2004	46°54.3′ N 91°54.2′ W	185	Duluth	19	1.5	Zea mays	1	64.8	-
	7/8/2004	44°14' N 95°19' W	350	Lamberton	3	1.2	Zea mays	1	60.8	-
Mississippi										

	7/18/2004	32°32′05″ N 90°20′76″ W	90	Jackson	32	1.8	Zea mays	2	62.8	3
Nebraska										
	7/30/2004	41°09′ 52″N 96°28′53″W	365	Mead	8	3	Zea mays	1	64.5	-
New Hampshire										
	7/27/2004	43°06′35″ N 70°56′55″ W	25	Durham	5	2	Zea mays	1	66.4	-
New Mexico										
	7/19/2004	35°41′13″ N 105°56′16″ W	1635	Santa Fe	32	0.6	Zea mays	3	67.5	3.6
New York										
	7/20/2004	42°43′52″ N 73°59′46″ W	115	Schenectady	24	0.9	Zea mays	2	61.8	2.1
North Carolina										
	~ 7/5/04	36°07′20″ N 78°41′10″ W	90	Creedmoor	8	1.8	Zea mays	1	59.9	-
Ohio										
	7/20/2004	39°41.019′ N 82°57.508′ W	205	Circleville	2	2.1	Zea mays	4	56.7	1.6
	8/4/2004	41°14′71″ N 81°36′25″ W	335	Kent	4	2.7	Zea mays	2	57.1	1.1
	7/20/2004	39°04.662′ N 83°00.603′ W	165	Piketon	2	1.5	Zea mays	2	54.4	0.1
	7/21/2004	39°58.806′ N 81°56.905′ W	255	Zanesville	8	1.2	Zea mays	1	56.3	-
Oklahoma										
	7/30/2004	35°04′080″ N 97°29′085″ W	350	Washington	1	1.5	Zea mays	2	62.9	0.3
Oregon										
	8/20/2004	44°01′35″ N 116°57′46″ W	655	Ontario	-	2	Zea mays	1	63.5	-
Pennsylvania										
	7/22/2004	40°26′26″ N 79°59′45″ W	235	Pittsburgh	64	1.2	Zea mays	1	53	-
	7/21/2004	40°02.040′ N 80°16.050′ W	300	Sparta	3	1.8	Zea mays	1	52.7	-
Tennessee										
	7/18/2004	35°18.990′ N 86°51.913′ W	240	Cornersville	2	1.8	Zea mays	1	64.6	-
	7/19/2004	36°28.429′ N 86°43.438′ W	235	White House	3	1.8	Zea mays	1	58.7	-
Texas										

	7/16/2004	31°27′07″ N 100°26′01″ W	565	San Angelo	24	1.5	Zea mays	2	67.7	1.3
Washington										
	8/19/2004	45°43′32″ N 121°49′05″ W	160	Carson	-	2.1	Zea mays	1	65.1	-
	8/8/2004	48°41′00″ N 122°54′00″ W	15	Eastsound	8	-	Zea mays	2	64.5	2.3
West Virginia										
	7/21/2004	39°37.673′ N 79°38.955′ W	480	Bruceton Mills	3	1.8	Zea mays	1	52.4	-
	7/20/2004	38°21.851′ N 82°28.357′ W	300	Huntington	3	1.7	Zea mays	1	54.2	-
	7/21/2004	40°00.841′ N 80°39.258′ W	205	Weeling	8	1.5	Zea mays	1	56.7	-
Wisconsin										
	7/3/2004	44°17′38″ N 90°50′52″ W	240	Melrose	10	1	Zea mays	1	64.8	-
	7/25/2004	43°03'48″ N 87°57'59″ W	195	Milwaukee	50	1.5	Zea mays	1	65.3	-
	7/20/2004	42°59'26″ N 89°31'59″ W	300	Verona	2	2.1	Zea mays	1	60.9	-
Virginia										
	7/23/2004	38°01.475′ N 77°28.909′ W	70	Ladysmith	2	2.1	Zea mays	3	60.3	4.3
International Sites:										
Brazil										
	7/24/2004	22°42'30″ S 47°38'01″ W	554	Sito	-	-	Zea mays	1	65.1	-
Canada										
	7/22/2004	49°44'30″ N 112°17'07″ W	815	Taber, A.B.	10	1.4 - 1.5	Zea mays	2	67.7	3.6
	8/30/2004	44°39' N 63°36' W	145	Halifax, N.S.	0.8 - 1.6	1.7	Zea mays	2	60.7	1.3
China										
	8/19/2004	38°16′09″ N 106°48′34″ E	1348	Yinchuan	-	-	Zea mays	1	59	-
Mexico										
	7/25/2004	19°57' N 103°46' W	-	Tapalpa	8	1.8	Zea mays	1	64.6	-
Venezuela										
	8/26/2004	9°23'33″ N 66°38'30″ W	-	El Sombrero	45	2	Zea mays	1	64.6	-

8/18/2004	closest - 10°21'71″ N 66°57'43″ W	-	Caracas	30	1.5	Zea mays	1	64	-

1. We used barley (FIRI G) as a secondary standard, and its standard deviation was 2.3‰ based on 13 replicates scattered across multiple wheels. The two other secondary standards we used were an oxalic acid (OX-II) and an Australian National University (ANU) standard. These had standard deviations of 2.0‰ (with 4 replicates) and 2.5‰ (with 4 replicates), respectively. Based on the accuracy of these three secondary standards, (FIRI G, OX-II and ANU) we assumed that the accuracy of any individual measurement was ± 2.3‰.

As a measure of precision, we also provide the standard deviation of samples that we ran multiple times (and on different AMS wheels). The pooled mean standard deviation across sites for which we made multiple measurements was 2.4‰.

A.1 Ancillary methods and results

We chose to use corn in our study because it is an annual plant that grows ubiquitously across North America. It has a relatively short growing season that allowed us to map out regional patterns of atmospheric radiocarbon over a period of a several months (May-July of 2004). Annual plants have a distinct advantage over perennial plants in terms of their use for this purpose because they accumulate all of their biomass within a single season – no corrections are required to account for carbon fixed in previous years.

To model the effect of cosmogenic production in the upper troposphere and stratosphere on $\Delta^{14}C$ gradients, we used six upper atmosphere pulse functions that represented production at two altitude levels (200 mb and 90 mb, corresponding roughly to the upper troposphere and the stratosphere) and three latitude bands (south of 30 °S, 30 ° S to 30 °N, and north of 30 °N). We distributed ^{14}C production equally between northern and southern hemispheres, with 60% at 200 mb and 40% at 90 mb, and with 70% at high latitudes and 30% at low latitudes. We chose this distribution to capture the pattern predicted from theory [*Jöckel et al.*, 1999; *Lingenfelter*, 1963]. We assumed that long-term cosmogenic production averages 6.6 kg ^{14}C yr^{-1}, roughly balancing decay in short-term carbon pools [*Goslar*, 2001] and consistent with observed ^{14}CO concentrations [*Quay et al.*, 2000].

As a sensitivity test, we simulated the terrestrial biosphere-induced gradient in $\Delta^{14}C$ across the continent for two cases where we doubled and halved the carbon residence times with the CASA model. Doubling the carbon residence times decreased the difference in surface air $\Delta^{14}C$ by 0.1‰ (from 0.4‰ to 0.3‰) between the eastern

U.S. and the mountain west regions and by 0.2‰ (from 0.9‰ to 0.7‰) between the Ohio/Maryland and the mountain west regions. Similarly, halving the carbon residence times decreased the difference in surface air $\Delta^{14}C$ by 0.2‰ and 0.3‰, respectively, for the same two sets of regions. This led us to conclude that our modeled gradient of $\Delta^{14}C$ in surface air caused by biosphere-atmosphere exchange was relatively insensitive to our representation of carbon cycling within the terrestrial biosphere model. It also strengthens our conclusion that most of the variability in surface air $\Delta^{14}C$ across North America is caused by fossil fuel emissions.

Ancillary References

Goslar, T. (2001), Absolute production of radiocarbon and the long-term trend of atmospheric radiocarbon, *Radiocarbon*, *43*, 743-749.

Jöckel, P., et al. (1999), Simulations of cosmogenic ^{14}CO using the three-dimensional atmospheric model MATCH: Effects of C-14 production distribution and the solar cycle, *J. Geophys. Res.*, *104*, 11733-11743.

Lingenfelter, R. E. (1963), Production of carbon-14 by cosmic-ray neutrons, *Rev. Geophys.*, *1*, 35-55.

Quay, P., et al. (2000), Atmospheric (CO)-C-14: A tracer of OH concentration and mixing rates, *J. Geophys. Res.*, *105*, 15147-15166.